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Techno-economic viability of innovative membrane systems in water and mass recovery from dairy wastewater



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ABSTRACT

Viability of innovative treatment systems in recovery of water and mass from dairy wastewater was techno-economically investigated. Lab-scale experiments were carried out for water recovery from whey wastewater using two combined membrane processes as forward osmosis (FO)/membrane distillation (MD) and MD/reverse osmosis (RO). Raw whey was concentrated to the solid contents of 21.0% and 25.8% by FO/MD and MD/RO, respectively. Production of commercial whey powders was successfully accomplished by spray drying of the concentrated whey streams. Full-scale costs of both systems were individually estimated using process modeling and cost estimation software. The simulations for a design influent of 100 m³/day showed that water can be recovered in sufficient quality to be reused in cheese production and the recovered amount increases up to 66–68% compared to 30% for that of UF/RO system. Besides, both treatment systems yielded a return of 12–13 million \$ with annual net profit of about 800,000 \$ as competing with UF/RO. Pay-back times of the system investments were determined as satisfactory as under 1 year due to annual revenues of about 3.4 million \$ from water recovery and whey powder selling. The innovative systems studied seemed to have conclusively enabled more sustainable dairy waste management with good economic benefits.

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1. Introduction

Dairy processing effluents are ranked among the major sources of industrial water pollution problems with 350,000 t of annual production in Turkey. Three common control technologies are utilized mainly in whey treatment: (i) valorization for recovery of valuable whey compounds, (ii) fermentation for production of lactic acid, ethanol or hydrogen, and (iii) physicochemical treatment such as thermal techniques, coagulation–flocculation, precipitation, electrochemical oxidation, membrane separation, etc. [1–3]. Whey may cause serious operating problems in traditional wastewater treatment plants and could lead to insufficient quality in effluents being discharged into the environment because of its extremely high organic and nutrient loads. Thus, the treatment of whey wastewater necessitates facilitating more effective and successful management strategies which may preferentially and reliably involve applications of innovative treatment technologies as being more environmentally benign.

In recent times, researchers shifted their interests in reuse or recycling of dairy wastewaters [4–6]. Significant developments in membrane technology have also increased the reuse capability and recycling extent of dairy wastewater [7]. In this context, various membrane processes were studied such as the membrane bioreactors [7,8], nanofiltration (NF) together with various treatment processes [9], ultrafiltration/nanofiltration (UF/NF) [10], reverse osmosis (RO) [5], and NF/RO [11]. In whey treatment, membrane processes were successfully employed for the concentration and demineralization of whey [12], the fractionation of whey proteins from lactose and salts [13], and the purification of lactose in the concentrated whey [14]. Despite the fact that RO process could provide high rejections of lactose, chemical oxygen demand (COD) and nutrient contents, its operation in single step is insufficient for producing the reuse water in a desired quality. Therefore providing an effective water recovery in addition to concentrating the raw whey using membrane processes becomes more prominent for clean production practices in cheese manufacturing. In that sense, the utilization of various membrane processes together in line with an integrated treatment strategy seems to have technological novelty to simultaneously reach targets comprising purifying water and producing whey powder from raw whey. These systems may include at least one innovative

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membrane process such as forward osmosis (FO) or membrane distillation (MD) that may be considered to provide low-cost treatability and more flexible operability in practice.

FO is an osmotic-driven process whereby a concentrated draw solution is placed on the other side of a semipermeable membrane to drive the transport of water molecules from a dilute feed solution to the concentrated draw solution. The driving force for the process is the osmotic pressure difference between feed solution (FS) and draw solution (DS). Thus it typically requires relatively and significantly less consumption of electrical energy in which the overall energy cost of FO can be much lower than pressure-driven membrane processes where the DS can be easily recovered or discarded using lower energy quality [15,16]. For this reason, FO must be coupled with other membrane techniques (like tight NF, RO and MD) to recover water from draw solutions in order to constitute its sustainable operation. Another innovative membrane process, MD is a thermally driven process, in which only vapor molecules are transported through membranes that prevent entering of bulk liquid solutions into the membrane's pores due to the surface tension forces. It has been used in many specific areas, yielding highly purified permeate and separating contaminants from liquid solutions, such as concentrating aqueous solution in fruit juices, pharmaceutical industry, wastewater treatment and water desalination [17,18]. Both innovative processes are to be able to confer favorable advantages due to their efficacy in operating conditions close to or at ambient temperature and pressure. So, on the eve of their becoming widespread in industrial activities, especially in dairy industry, remarkable case studies including specific recoveries of both water and concentrated valuable ingredients could be developed without notable product differentiations or deteriorations.

This work aims to investigate the techno-economic analysis of whey concentration and water recovery using FO/MD and MD/RO combined systems. Technical performances of the systems were experimentally determined by continuous operation of the processes in sequential mode. Based upon simulation of the technical performances in the process modeling and cost estimation software, a full economic analysis was carried out for the whole treatment systems complemented with a whey powder production line. Thus, real-world feasibilities of innovative membrane systems in whey management were revealed by comprehensively evaluating the simulated technical and economic results together with those of a typical, widely used UF/RO system [19].

2. Materials and methods

2.1. Materials

Acid cheese whey was supplied from industrial facilities of Cayirova Milk&Milk Products Inc., located at Kocaeli, Turkey (Table 1). FO and RO experiments were executed using flat sheet membranes, CTA (Hydration Technologies Inc., OR) and LFC-3 (Hydranautics Inc., CA), respectively. MD membranes were selected as polypropylene (PP) and polyvinylidene fluoride (PVDF) membranes having 0.22 μm pore size (GE Osmonics, MN). Analytical grade chemicals were obtained from Merck.

2.2. Experimental setups

2.2.1. FO/MD membrane system

A lab-scale FO process shown in Fig. 1(a) was operated with a cross-flow FO module which was a custom made cell with equivalent flow channel at both sides of the membrane. The module made from Deldrin acetal resin material (DuPont, Delaware) has an effective membrane area of 140 cm^2 . Two speed controllable peristaltic pumps (EW 77111-67, Cole Parmer, IL) were used to pump the solutions.

Table 1
Characteristics of raw whey solutions used.

Parameters	Unit	Raw whey (A) [*]	Raw whey (B) [*]
pH	–	4.75	5.17
E_c	$\mu\text{S}/\text{cm}$	7890	9640
Cl^-	mg/L	1355	1250
Osmolality	mmol/kg	314	396
Density	g/cm^3	1.0213	1.018
SCOD	mg/L	59,280	63,034
$\text{NH}_4\text{-N}$	mg/L	113	101
$\text{NO}_2\text{-N}$	mg/L	0.13	0.12
$\text{NO}_3\text{-N}$	mg/L	149	119
TKN	mg/L	1159	1325
TN	mg/L	1308	1445
$\text{PO}_4\text{-P}$	mg/L	363	385
TP	mg/L	455	483
Total protein	%	2.30	2.87
Fat	%	0.19	0.21
SNF	%	6.58	8.15
Total solid content	%	6.77	8.36
Lactose	%	3.56	4.26
Minerals	%	1.25	1.65

^{*}Raw whey (A) and (B) were the whey solutions used as the feed streams in “FO/MD” and “MD (PVDF)/RO”, and “MD (PP)/RO” integrated membrane systems, respectively.

Two flow meters were separately placed on the feed and draw lines of the setup to enable the desired flow rates on each line. The setup was also equipped with two constant temperature water baths (462-7028, VWR Scientific, IL) to maintain the same temperature at both the feed and draw solutions during the FO tests. Throughout the experiments, the salt concentration in the draw was kept constant. MD experiments were also carried out using the system depicted in Fig. 1(a). The process was operated under direct contact conditions using a separate MD module having same properties with FO module. Yet, water baths were separately adjusted in order to obtain temperature-driven operation by heating and cooling of feed and filtrate streams, respectively.

2.2.2. MD/RO membrane system

MD experiments in MD/RO combined system were executed using the same MD process represented in Fig. 1(a) and two experiments using PVDF and PP membranes were done under the same process operating conditions so as to determine the technical performances in the case of different membranes, due to their relatively superior performances versus inorganic and organic solutions, respectively [20,21]. RO experiments were operated using pressure-driven membrane system shown in Fig. 1(b). The system had a flat-sheet cross-flow membrane module having an effective membrane area of 140 cm^2 (GE Osmonics, MN). It was equipped with a feed tank, high-pressure pump of 100 bar with a flow volume 330 L/h (Bosch, Germany), flow splitter, digital flow meter (max 720 L/h) (Honsberg, Germany), and manual oil pump for clamping the module. The experiments were employed in the concentration mode, which means that permeate solutions were not returned into the feed tank while the concentrate was. The flow rates of the permeate solutions collected in a beaker were measured by an electronic balance (Precisa XT2220M-DR) and recorded by a computer.

2.3. Operating procedures

2.3.1. FO/MD membrane system

The FO/MD system included concentrating the raw whey by FO in the first step, and then processing the FO draw solution by MD in the second step. FO process was operated with 2 M NaCl draw solution at the conditions of feed and draw volumes of 3.5 L, cross-flow rate of 300 L/h (0.5 m/s), temperature of 25 ± 0.5 $^\circ\text{C}$, reverse membrane

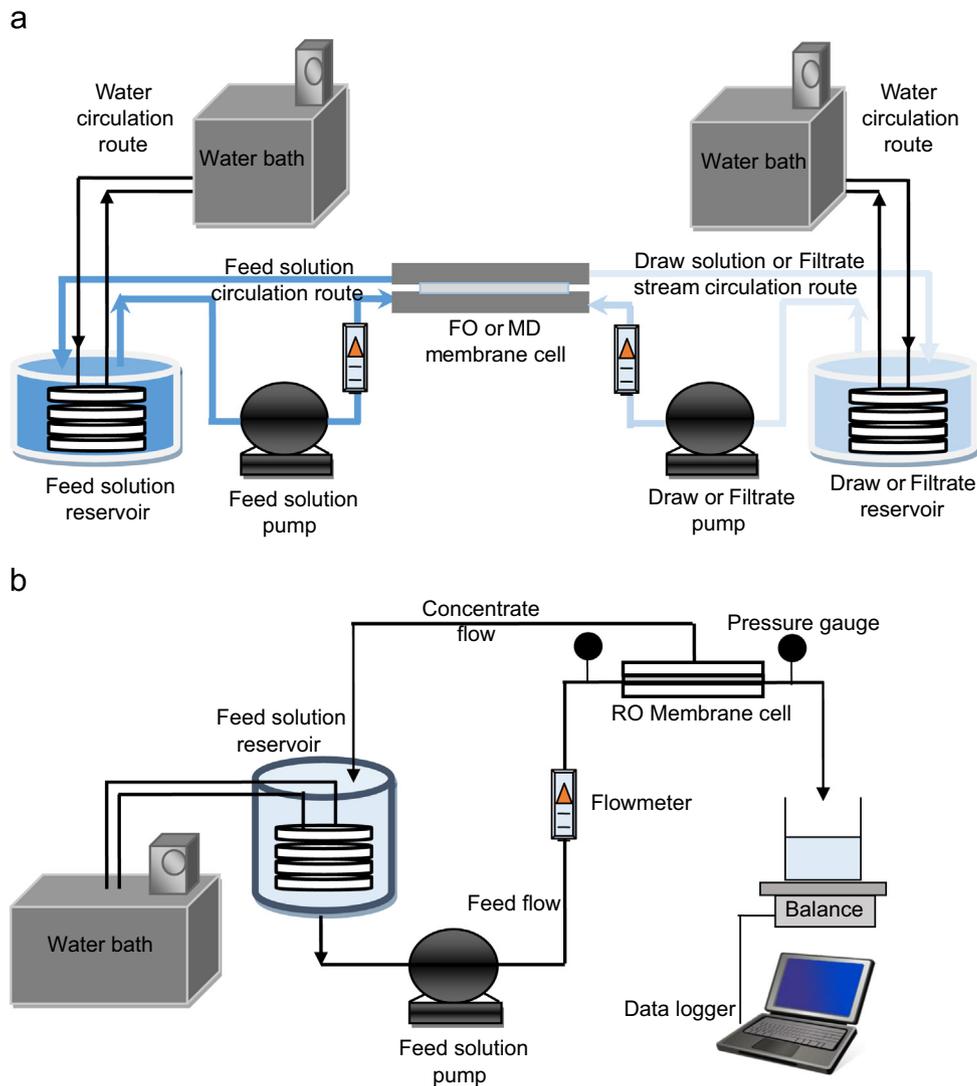


Fig. 1. Experimental FO or MD (a) and RO (b) setups.

orientation mode (the active layer in contact with feed whey) and co-current flows in the channels. The process was conducted at 4 consecutive periods in a total duration of 21 h. At the end of operating periods, the fouled membrane was subjected to a cleaning of 15 min using de-ionized water and fresh 2 M NaCl as the feed and draw solutions, respectively, after turning the membrane to the normal mode.

The MD process was employed using hydrophobic PVDF membrane. The temperature values were set as 48 ± 1 and 20 ± 1 °C for feed and filtrate streams, respectively, and thus the process was operated at a constant transition temperature difference of 28 ± 1 °C through the membrane. At the same starting volumes of 3.25 L at both sides, 18-h operating in total duration was carried out at 3 consecutive periods. 17-mil spacer was used at the flow channels of the module in order to achieve turbulent flow conditions (Reynolds number ≈ 3300 –4500). For this, the experiments were conducted at the cross flow rate of 180 L/h in both channels. The treated membrane was merely kept in distilled water at room temperature between operating runs, without applying any special membrane cleaning procedure.

2.3.2. MD/RO membrane system

In the MD/RO system, first the raw whey was concentrated using a direct contact MD process, and then MD filtrate was

processed by RO to produce clean water. The hydrophobic PP and LFC-3 membranes were used in MD and RO processes, respectively. Using the same starting volumes, operating temperature and hydrodynamic flows as with MD in the FO/MD system, MD experiments in this combined system were executed with entire periods of 13 h and 10 h for PVDF and PP membranes, respectively, due to their different efficiencies in concentrating raw whey. RO process was applied with 2 L feed volume, at 35 ± 0.5 °C temperature, 2.5 m/s cross-flow rate, 40 bar transmembrane pressure and for 2.2-h duration.

2.4. Analytical methods and process performances

Analytical procedures for the measurement of water quality parameters in raw whey, concentrated whey, draw solution and permeate streams of combined systems were defined in Aydinler et al. [22]. Qualities of whey powders were analyzed in dairy laboratories of the Scientific and Technological Research Council of Turkey (TUBITAK) accredited by the European co-operation for Accreditation (EA) and the International Laboratory Accreditation Cooperation (ILAC). The principles on the determination of technical performances of FO and RO processes were given in Aydinler et al. [19]. MD performance calculations for rejections and permeate flux were carried out in the same manner as in RO process calculations.

In FO process, osmotically-driven force is the net osmotic pressure difference ($\Delta\pi_{\text{net}}$) which refers to the osmotic pressure difference between the draw and feed solutions. It was determined from the difference of the osmotic pressures determined for both solutions after osmolality measurements in the sample solutions by using the following equation:

$$\Delta\pi_{\text{net}} = \pi_d - \pi_f \quad (1)$$

where π_d and π_f are the osmotic pressures of the draw and feed solutions, respectively. The osmotic pressures of the solutions were calculated in accordance with the van't Hoff equation.

$$\pi = RT[md] \quad (2)$$

where R is the ideal gas constant (8.314 J/K·mol), T the absolute temperature (K), m the osmolality (mosm/kg), and d the solution density (kg/L). It should be noted that the multiplication term in square bracket is referred to as the osmolarity (mosm/L) representing the total solute concentration in the solution.

2.5. Economic analysis of combined treatment systems

In addition to the experimental trials for technical performance evaluation, efforts were placed on conducting a comprehensive economic analysis to estimate the possible costs and benefits that would arise from the real-scale applications of the systems. For this, each innovative membrane system (FO/MD and MD/RO) was coupled with a conventional whey production line. On the other hand, to make a detailed comparison toward field practices of innovative systems, economic investigation of UF/RO system was additionally carried out by directly making a new simulation using its technical performances simulated in the software formerly used in Ref. [3].

The economic feasibilities of the final combined systems were carried out using process design software equipped with a cost estimation module (Intelligen's *SuperPro Designer*[®] v7.5). In the first step of economic analyses, material balances of three membrane systems at a design inflow of 100 m³ per day were individually calculated by using the resultant technical performances obtained from lab-scale treatments. The design permeate fluxes of the processes were 4.0/4.0 L/m² h and 7.3/40.0 L/m² h for FO/MD and MD/RO systems, respectively. They were selected as 10.3/33.4/40.0 L/m² h for UF/RO (first step RO)/RO (second step RO), as presented in Ref. [3]. For all the processes in the systems, any membrane cleaning frequency was taken into account in terms of their application into the practice. Hence membrane cleaning costs were neglected in the economic analyses of the systems. On the other hand, membrane areas were assumed according to the maximum limit of 40 m² per the module, as commonly applied in the practice. For low membrane areas required in the process designs, continuous reliable operability was basically considered for the situations such as maintenance, cleaning and offline.

After several runs of the program, the process models managed to verify both the experimental results, and thereafter predefined design limitations for dimensioning of the system units. At the next step, the economic evaluation module of *SuperPro Designer* was adjusted to local prices and other economic conditions. The individual prices associated with the equipment, consumables (NaCl), utilities (electricity and heat agents), disposals (wastewater and membranes used) and pure components (water and whey powder) were retrieved from a detailed research in both literature and the local market. Also labor costs were estimated by considering local hourly wages and working hours limits for 7920 h of annual operation (For details see Ref. [3]). While the total capital cost estimation includes direct fixed capital, contractor's fee and contingency, working capital and start-up costs calculations, annual total operating costs were separately estimated by the software based on

labor, facility, consumable, disposal and utility-dependent costs. In addition, the total revenue to be gained from the real-scale operation was estimated from unit revenues from recycled water (1.8 \$/m³ water) and whey powder selling (32.5 \$/package of 25 kg). The other economic parameters as return on investment, payback time and net present value (NPV) were determined by the cash flow analysis conducted in the software at the base of 3 year loans at a fixed interest rate for 15-year plant life. More details about extensive examination of the economic analysis procedure of the software can be found in Vergili et al. [23].

For the scope of economic investigation, a special concern was given to the estimation of membrane purchase costs. RO membrane purchase cost of \$30 per m² was obtained from the Turkey branch office of an international membrane company. Whereas FO and MD membrane prices were estimated from lumped module purchase and construction costs due to lack of knowledge on their market prices. In accordance with the previous literature studies, the FO membrane price assumed as \$12 per m² of membrane corresponded to \$55–60 per m² of membrane in FO module as a result of \$1000 module purchase cost, \$500 module construction cost and other indirect costs [3]. Also, the typical microfiltration membrane price (\$100/m² of membrane) for the MD membrane was considered that excludes the membrane module purchase, construction and other indirect costs due to them being the same prices as for FO process [23].

3. Results and discussions

3.1. Technical performance results

3.1.1. FO/MD combined system

In FO/MD whey treatment system, technical performances of the processes on whey concentration and clean water production were investigated by monitoring variations in the parameters as volumetric water permeations (FO and MD), water fluxes (FO and MD), salt flux (FO), osmotic pressures of solutions and net osmotic pressure difference (FO), and whey solid content (FO) with time. The results are all shown in Fig. 2(a)–(e).

It can be seen from Fig. 2(a) that while 2.4 L of water in raw whey passed into draw solution in the FO process, 1.7 L of water of FO draw solution was transported to the MD permeate stream. The FO water flux prominently decreased along filtration time and operating periods in spite of membrane cleaning carried out by back-washing. Besides, noticeable flux differentiations in MD were not observed, especially for the second and the third periods (Fig. 2(b)). The last FO water fluxes were determined as 9.0, 8.3, 3.0 and 3.9 L/m² h in increasing order of operating periods (6, 12, 18, and 21 h), respectively. At the end of each backwash cycle, the initial water flux of each period did not reach the initial value of the previous one. Especially at the third period, a flux decline was determined to occur at a significant level. It could be therefore said that in practice, in order to operate the process continuously at a high water flux, membrane back-washing process should be done at least by increasing the frequency with time. Otherwise, efficient cleaning procedures should be developed during pilot-scale investigations. On the other hand, the last MD water fluxes were established as 6.3, 4.2 and 4.0 L/m² h at the end of the periods I, II, and III, respectively. The changes in water flux showed that clean water production by MD process could be made sustainable at much more long-term operation, when compared to flux deteriorations in the FO process.

Fig. 2(c) indicates that the variations of periodic salt fluxes in the FO process performed salt transitions having a maximum peak with increasing values in shorter time along increasing periods. The last salt fluxes were found as 0.40, 1.63, 0.45 and 1.35 g/m² h

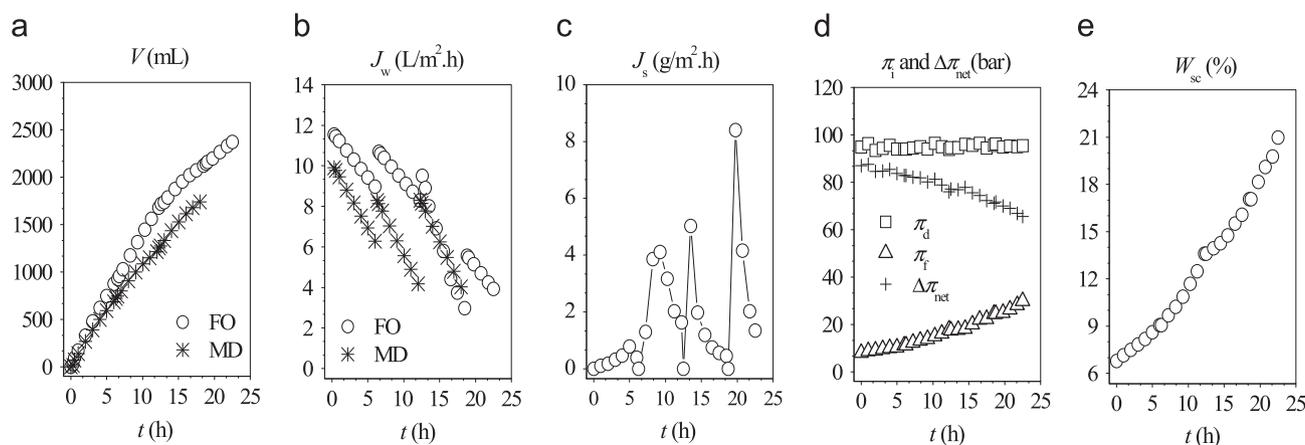


Fig. 2. Technical performances of FO and MD processes respectively on whey concentration and clean water production in FO/MD membrane system ((a) volumetric water permeations in FO and MD; (b) water fluxes in FO and MD; (c) salt flux in FO; (d) osmotic pressures of solutions and net osmotic pressure difference in FO; and (e) whey solid content in FO) (FO–CTA membrane, 3.5 L volumes in feed and draw, 2 M NaCl draw solution, 21 h duration, 300 L/h cross-flow rate, 25 ± 0.5 °C temperature, reverse mode and co-currently flow in FO; hydrophobic PVDF membrane, 3.25 L volumes in feed and filtrate, 18 h duration, 180 L/h cross-flow rate, 48 ± 1 and 20 ± 1 °C temperatures in feed and filtrate, 28 ± 1 °C transition temperature difference in MD).

Table 2
Technical performances of FO and MD processes in FO/MD system and quality of recovery water produced by MD process.

Parameter	Unit	FO/MD system				
		FO process		MD process		
		Concentrated whey	^a 2 M NaCl draw	^b Concentrated feed	Permeate	R (%)
pH	–	5.0	4.6	6.7	7.2	–
E_c	$\mu\text{S/cm}$	2560	157,900	316,000	45.0	99.99
Cl^-	mg/L	7148	66,479	117,564	12.0	99.99
Osmolality	mmol/kg	1140	3736	9708	5.0	99.95
Density	g/cm^3	1.058	1.062	1.123	0.997	11.22
COD	mg/L	140,348	1739	5138	38.0	99.25
$\text{NO}_2\text{-N}$	mg/L	0.37	0.17	0.37	0.00	100.00
$\text{NO}_3\text{-N}$	mg/L	310	8.8	18.9	0.03	99.86
TKN	mg/L	2760	25.8	43	18.2	57.88
TN	mg/L	3070	34.8	62	18.2	70.82
$\text{PO}_4\text{-P}$	mg/L	736	0.6	1.3	0.00	100.00
TP	mg/L	891	3.7	2.0	0.00	100.00

^a At the beginning of the experiment, FO draw solution had a water quality of 6.25 pH, 156.7 mS/cm conductivity, 66,080 $\text{mg Cl}^-/\text{L}$, 3679 mmol/kg osmolality and 1.060 g/cm^3 density.

^b Because 2 M NaCl draw operated in FO was used as the MD feed; concentrated feed stream in MD means the FO draw concentrated by MD processing.

for operating periods of 6, 12, 18, and 21 h, respectively. According to the variations of the osmotic pressure of the feed and draw solutions and the net osmotic pressure difference presented in Fig. 2(d), the process was operated with a constant osmotic pressure of about 95 bar in the draw by continuous addition of NaCl during the filtration. But, the influence of net drive force on water transport decreased with time due to salt leakages into the whey. As a significant indicator for whey concentration, the last whey solid contents reached 9.1%, 13.6%, 17.1% and 21.0% for operating periods I, II, III, and IV, respectively (Fig. 2(e)). The results obtained for FO/MD integrated system showed that an effective concentration of cheese whey can be provided by withdrawing of water from whey into the FO draw solution. But, the MD performance is the main distinctive feature for clean water production. In FO/MD system, technical performances of both membrane processes were presented in Table 2, together with the quality features of clean water produced. While the conductivity value in the FO draw solution increased from 157.9 mS/cm to 316.0 mS/cm, it was measured to be 45 $\mu\text{S/cm}$ in the MD permeate stream. The osmolality of the FO draw increased from 3736 mmol/kg to 9708 mmol/kg, while that of the MD permeate was 5 mmol/kg. Despite high chloride and COD

concentrations in the influent, very low values of 12.0 and 38.0 mg/L were measured in the effluent, respectively. Moreover none of $\text{NO}_2\text{-N}$, $\text{PO}_4\text{-P}$ and TP compounds were observed in the MD permeate stream, and also the concentration of $\text{NO}_3\text{-N}$ was very low at a value of 0.03 mg/L. However, a relatively high nitrogen concentration of 18.2 mg/L was determined in the permeate stream of which a significant part was in the form of organic nitrogen. As a consequence, it was deduced that in the water recovery-oriented use from whey of FO/MD combined system, the production of high quality water having rather low conductivity at about neutral pH was proved accompanied with rather good performance of the system for whey concentration. So, the resulting water could be reused in the cheese production process or any other suitable purpose at industry scale.

3.1.2. MD/RO combined system

In MD/RO whey treatment system, technical performances of the processes on whey concentration and clean water production were explored by the time variations of the parameters as water

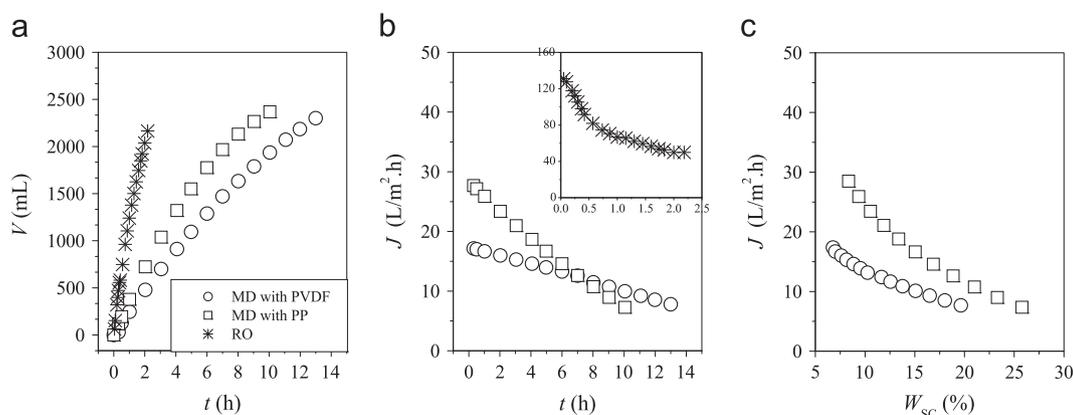


Fig. 3. Technical performances of MD and RO processes respectively on whey concentration and clean water production in MD/RO membrane system ((a), water volume; (b) permeate flux vs. time (same operating conditions with MD in FO/MD except for durations of 13 and 10 h for hydrophobic PVDF and PP membranes in MD); LFC-3 membrane, 2 L feed volume, 2.2 h duration, 2.5 m/s cross-flow rate, 35 ± 0.5 °C temperature, 40 bar trans-membrane pressure in RO); and (c) permeate flux vs. whey solid content.

Table 3

Technical performances of MD and RO processes in MD/RO system and quality of recovery water produced by RO process.

Parameter	Unit	MD/RO system								
		MD Process (PVDF membrane)			MD Process (PP membrane)			RO Process ^a		
		Concentrated whey	Permeate	R (%)	Concentrated whey	Permeate	R (%)	Concentrated feed ^b	Permeate	R (%)
pH	–	4.6	5.0	–	4.9	5.3	–	5.3	5.4	–
E_c	$\mu\text{S}/\text{cm}$	16,300	330	98.00	17,400	147	99.15	168	18.3	89.14
Cl^-	mg/L	3572	60	98.32	4549	12.5	99.73	14.2	0.0	100.00
Osmolality	mmol/kg	1085	10	99.08	1143	2.0	99.83	1.0	0.0	100.00
Density	g/cm^3	1.060	0.998	5.82	1.059	0.998	5.82	0.990	0.994	–0.40
COD	mg/L	139,748	804	99.42	153,733	176	99.89	313	17.1	94.56
$\text{NO}_2\text{-N}$	mg/L	0.40	0.00	100.00	0.49	0.01	97.57	0.10	0.00	100.00
$\text{NO}_3\text{-N}$	mg/L	310	2.1	99.32	487	0.8	99.83	1.7	0.0	100.00
TKN	mg/L	3705	17.4	99.53	3531	22.4	99.37	24.6	18.5 ^c	25.00
TN	mg/L	4015	19.5	99.51	4018	23.3	99.42	26.4	18.5 ^c	29.99
$\text{PO}_4\text{-P}$	mg/L	687	4.9	99.28	688	0.9	99.87	1.0	0.0	100.00
TP	mg/L	956	6.5	99.32	1065	1.9	99.82	2.2	0.0	100.00

^a RO process was applied to MD process with PP membrane due to its better performance in clean water production in the system [20].

^b Concentrated feed stream in RO process was the MD permeate stream concentrated by RO processing.

^c $\text{NH}_4\text{-N}$ in RO permeate was measured as 0.06 mg/L which means that nitrogen compounds passing to the permeate stream originated mainly from organic nitrogen.

volume and permeate fluxes, and the results were depicted in Fig. 3(a) and (b), respectively.

As can be seen from Fig. 3(a), in the first step of the combined system, 2.3 and 2.4 L of water of raw whey were withdrawn into the MD permeate stream using PVDF and PP membranes, respectively. In the second step of the system, the permeate volume in RO process was about 2.2 L. The variations of the water flux values of the processes with the time as shown in Fig. 3(b) yielded that in the MD (PVDF) process, the initial permeate flux of $17.1 \text{ L}/\text{m}^2 \text{ h}$ decreased with increasing time to the last flux of $7.8 \text{ L}/\text{m}^2 \text{ h}$. On the other hand, the water flux in the MD (PP) process was also observed to decrease with time in which the initial permeate flux of $27.7 \text{ L}/\text{m}^2 \text{ h}$ decreased to the last flux of $7.3 \text{ L}/\text{m}^2 \text{ h}$. Despite the last and partially less permeate flux; the MD (PP) exhibited the same permeation performance in the entire time of 10 h, which is 3 h less than that of the MD (PVDF) (Fig. 3(b) and (c)). After the MD processing, the MD (PP) permeate stream was filtrated using RO which in turn produced clean water with a RO permeate flux as high as $50 \text{ L}/\text{m}^2 \text{ h}$ (Fig. 3(b)). The last water fluxes during the raw whey processing with the MD processes with PVDF or PP membranes in MD/RO were higher than the last water fluxes of 3.0 and $4.0 \text{ L}/\text{m}^2 \text{ h}$ in the FO and MD in FO/MD system, respectively. These

results deduced that although single MD process without complementary treatment may be mostly preferred in practice, appropriate operating conditions concerning backwashing and working procedures have to be particularly examined prior to field applications. More importantly, its single practice during whey processing can be entirely valid only in case of obtaining an effluent with the desired quality.

In MD/RO system, technical performances of both membrane processes were presented in Table 3, together with the quality features of clean water produced. High amounts of organic contaminants in whey led to somewhat high permeate concentrations in spite of high rejection performances in the process. In the permeate streams of the MD operated with PVDF and PP membranes, COD, TP and TN parameters were measured as 804 and 176 mg/L, 6.5 and 1.9 mg/L, and 19.5 and 23.3 mg/L, respectively. Despite that, relatively low effluent conductivities were determined with 330 and $147 \mu\text{S}/\text{cm}$ for PVDF and PP membranes, respectively. These results indicated that on the one hand the raw whey can be concentrated with the direct contact MD process at satisfactory levels, while on the other hand the treated water in high quality could not be produced by MD alone. However the production of clean water can be made possible by using an innovative MD/RO

combined system. It can be seen from Table 3 that the RO effluent had suitable quality for reuse in cheese production. COD and TN were as low as 17.1 and 18.5 mg/L, respectively, with a very low conductivity of 18.3 $\mu\text{S}/\text{cm}$ at weak acidic pH of 5.4, while nitrite, nitrate, orthophosphate, TP and chloride were not present in effluent. These results proved that MD/RO produced somewhat better effluent quality when compared with that of FO/MD.

3.1.3. Quality contents of whey concentrated by combined systems

Quality contents of the whey concentrated by FO/MD and MD/RO combined systems were analyzed and time-dependent variations of whey parameters as fat, SNF, total solid, total protein, lactose and minerals contents are indicated in Fig. 4(a)–(c).

By continuous operations of FO, MD (PVDF) and MD (PP) processes, the concentrated whey solutions with total solids of 21.0%, 19.6% and 25.8% were achieved at the end of the total

durations of 21, 13 and 10 h, respectively, in which significant losses in raw whey ingredients were not observed during each filtration experiment due to stable or similar variations of all whey contents vs. time. According to these results, not only MD (PP) in the innovative MD/RO system exhibited the best whey concentration performance, but also the performances of other processes studied as FO in FO/MD and MD (PVDF) in MD/RO seemed to be rather good and rational when compared to an anticipated limit of 15–20% in UF/RO for the solid content of the processed whey [24].

3.1.4. Quality of whey powders produced by spray drying

The quality analysis results of whey powders obtained from drying of the concentrated whey solutions are shown in Table 4, together with the quality values of commercial whey powder quoted from the literature [1,25].

According to Table 4, parametric values of pH, total dry matter, moisture and minerals were found as similar in whey powders dried after FO and MD processing in FO/MD and MD/RO combined systems, respectively. The protein, ash and lactic acid values of whey powder of FO/MD were higher than those of MD/RO. However, the lactose and fat values of whey powder of MD/RO was higher than those of FO/MD. Besides relatively higher ash content in whey powder of FO/MD was due to salt transportation from the draw into the raw whey concentrated by FO. Consequently, whey powders produced from the concentrated whey streams of FO/MD and MD/RO seemed to have commercial value as compatible with the components values contained in the literature.

3.2. Economic performance results

Process flow charts for whey treatment systems comprising FO/MD and MD/RO membrane systems complemented with whey powder production line are represented in Figs. 5 and 6, respectively. Numbers and dimensions of the process units needed to be used in the systems were determined by verifying all of experimental performance results of both studied systems in the process modeling step of the software. The dimensioning results belonging to the systems' units are also given in Table 5.

As can be obviously seen from Figs. 5 and 6 the studied treatment scenarios cover three main process lines as performance indicators: (I) water recovery line on main effluent stream of the combined membrane systems, (II) centralized wastewater treatment (CWT) line originated from condensation of drying vapor of concentrated whey in which it was assumed that air was used in spray drying and hot air emissions were released into the

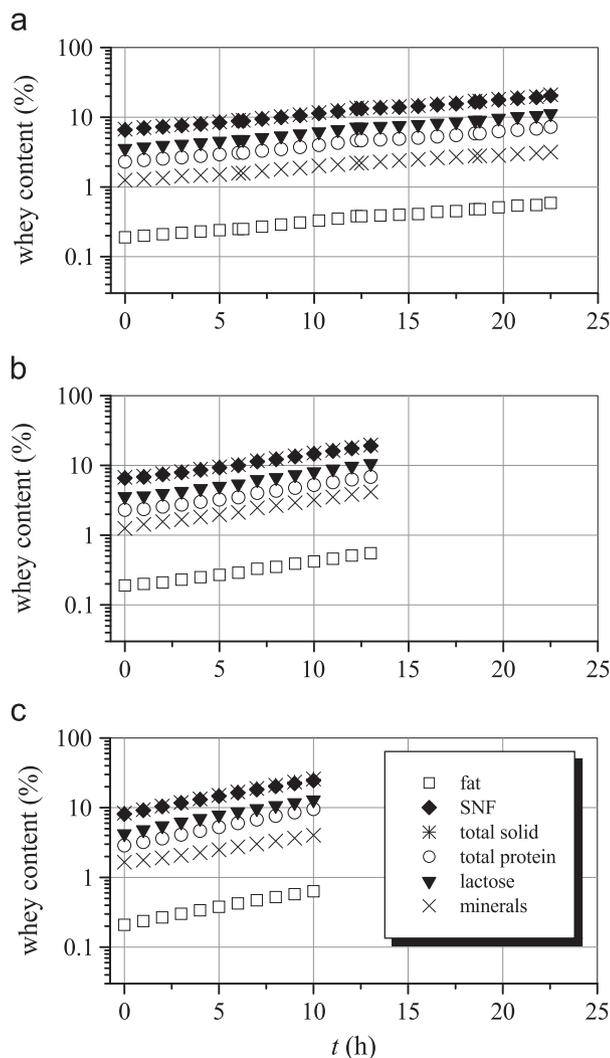


Fig. 4. Time-dependent variations of whey contents concentrated by FO process in (a) FO/MD system (FO-CTA membrane, 3.5 L volumes in feed and draw, 2 M NaCl draw solution, 21 h duration, 300 L/h cross-flow rate, 25 ± 0.5 °C temperature, reverse mode and co-currently flow in FO; hydrophobic PVDF membrane, 3.25 L volumes in feed and filtrate, 18 h duration, 180 L/h cross-flow rate, 48 ± 1 and 20 ± 1 °C temperatures in feed and filtrate, 28 ± 1 °C transition temperature difference in MD), (b) MD process with PVDF membrane in MD/RO system, (c) MD process with PP membrane in MD/RO system (same operating conditions with MD in FO/MD except for durations of 13 and 10 h for hydrophobic PVDF and PP membranes in MD; LFC-3 membrane, 2 L feed volume, 2.2 h duration, 2.5 m/s cross-flow rate, 35 ± 0.5 °C temperature, 40 bar trans-membrane pressure in RO).

Table 4

Chemical compositions of spray-dried acid whey powders produced from whey streams concentrated by FO/MD and MD/RO membrane systems.

Chemical composition	Unit	Literature values		FO/MD	MD/RO
		Ref. [1]	Ref. [25]		
pH	–	4.0–6.0	4.5–5.0	5.44	5.62
Total dry matter	%	94.0–98.0	95.0–96.5	95.11	95.49
Moisture	%	2.0–6.0	3.5–5.0	4.89	4.51
Total protein	%	10.0–14.0	9.0–12.0	14.02	11.99
Lactose	%	57.0–65.0	61.0–75.0	53.36	57.85
Fat	%	0.5–2.0	0.5–1.5	1.86	5.36
Ash	%	11.0–14.0	10.0–13.0	13.91	9.12
Minerals	%	0.7–10	–	2.19	2.46
Lactic acid	%	–	–	9.77	8.71

*Means analyses results which belong to whey samples obtained after spray drying of whey streams concentrated by the FO and MD (PP) processes in the FO/MD and MD/RO systems, respectively.

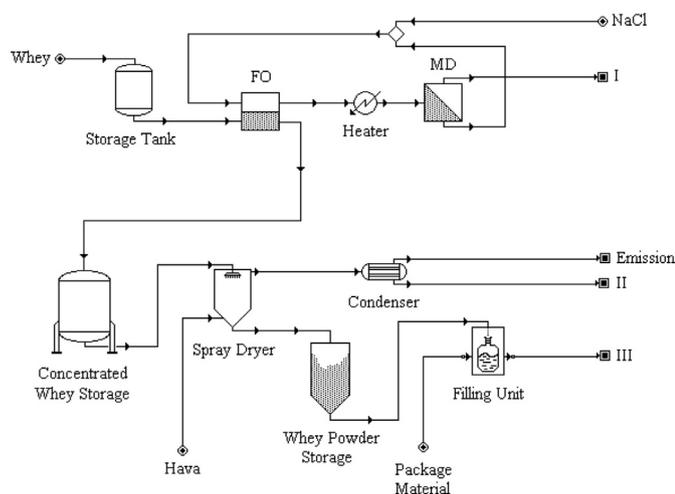


Fig. 5. Process flow sheets for FO/MD integrated membrane system accompanying with whey powder production line ((I) water recovery line, (II) centralized wastewater treatment line, and (III) packaged whey powder product line).

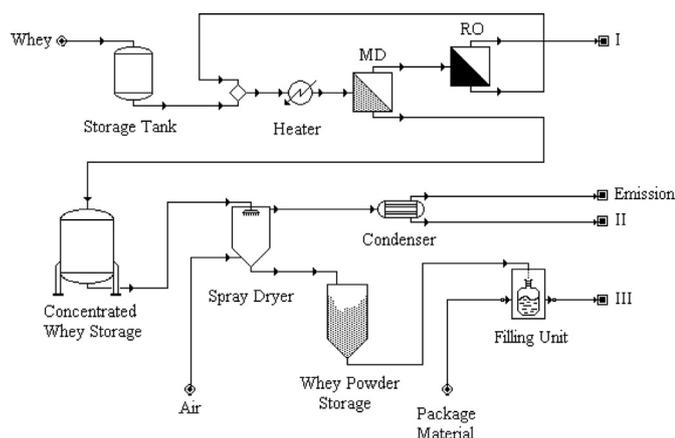


Fig. 6. Process flow sheets for MD/RO integrated membrane system accompanying with whey powder production line ((I) water recovery line, (II) centralized wastewater treatment line, and (III) packaged whey powder product line).

atmosphere after condensation, while condensation liquids were considered to be treated in CWT facility, and (III) packaged whey powder production line, which means the filling of the dried whey powder to 25 kg package entities.

The estimated simulative performances of the systems for continuous operating are shown in Table 6, accompanied with whey powder quality simulated for solid content, moisture and NaCl. The percent water recoveries in FO/MD, MD (PVDF)/RO, and MD (PP)/RO were determined as high as 67.3, 65.6, and 67.6, respectively, as 2.2–2.3 times greater than that of UF/RO (29.8%). In addition, the water quality in recovery line of FO/MD was to some extent better than that of UF/RO. Although both the studied systems allowed recovering water of good quality, the best water quality was obtained by MD (PP)/RO. In addition, no significant changes were determined for contaminant concentrations of wastewaters needed to be treated in a CWT facility. But, the decrease of flow rates in CWT lines of both systems compared to UF/RO brought about an advantage that enables the operation of the facility with less waste loads at the same conditions. Despite the fact that salt transition into the concentrated whey during FO processing in FO/MD led to a slightly higher NaCl amount in its whey powder, almost insignificant differences in the qualities of whey powders to be produced were found. It is found that the increase in water recovery performance of an integrated membrane system in whey treatment broadly resulted in an improvement in both mass loads of wastewater to be disposed of and whey product quality.

In light of operating cost, capital cost, revenues, return on investment, pay-back time and NPV parameters, overall economic outcomes were estimated separately for each whey treatment system based on different energy sources (electricity and waste heat) to be used in MD (Table 7). UF/RO, FO/MD and MD/RO enabled to produce whey powder with a production rate of about 103,000–104,000 entity/year for a whey inflow of 100 m³/day (330,000 m³/year). Unit production cost of whey powder of 4.23 \$/entity in UF/RO appeared to be increased up to around 5.98, 4.88 and 4.93 \$ in FO/MD, MD (PVDF)/RO and MD (PP)/RO, respectively. However, the use of waste heat instead of electricity in MD decreased the unit production costs to the values of 4.69, 3.64 and 3.74 \$/entity at the mentioned order. It can also be said that by considering high total revenue costs in the range about 3,350,000–3,400,000 \$/year, each system exhibited reasonable operating and capital investment costs. Pay-back time was under 1 year for all the systems in which time of 0.84 year for traditional membrane system

Table 5
Numbers and dimensions of process units used in whey treatment systems.

Unit process	Dimension	^a Number (membrane area required)			
		UF/RO	FO/MD	MD/RO	
				MD-PVDF	MD-PP
UF ^a	–	31 (9.73)	–	–	–
RO in the first step ^a	–	8 (9.34)	–	2 (27.35)	2 (28.20)
RO in the second step ^a	–	4 (7.76)	–	–	–
FO ^a	–	–	23 (39.73)	–	–
MD ^a	–	–	18 (39.31)	12 (39.28)	14 (37.20)
Raw whey storage	(2.5 m ³)	2	2	2	2
Concentrated whey storage	(1.5 m ³)	3	3	3	3
Spray dryer	(3.5 m ³)	4	3	3	3
Silo	(1 m ³)	2	2	2	3
Packaging	(7 pack/h)	2	2	2	3
Condenser	(15 m ²) ^b	3	2	2	2
Heater	(1 m ²) ^b	–	2	2	2

^a Those in parenthesis are the required membrane areas (m²) belonging to the corresponding membrane process in the combined system of interest (For example: 31 (9.73) for UF membrane process in UF/RO system means that totally 31 membrane modules having membrane area of 9.73 m² per module are required for UF process in the system).

^b Means the maximum heat transfer area and heat transfer area for the condenser and plate/frame heater units in the treatment systems, respectively.

Table 6
Simulated performances of whey treatment scenarios for flow rates and water qualities in raw whey line (RWL), water recovery line (WRL) and centralized wastewater treatment line (CWTL), together with simulated qualities for whey powder (WP).

Parameter	Unit	UF/RO				FO/MD				MD (PVDF)/RO				MD (PP)/RO			
		RWL	WRL	CWTL	WP	RWL	WRL	CWTL	WP	RWL	WRL	CWTL	WP	RWL	WRL	CWTL	WP
Flow rates	m ³ /day (kg/day) ^a	100.00	29.81	62.20	(7836) ^a	100.00	67.31	24.77	(7932) ^a	100.00	65.58	26.48	(7856) ^a	100.00	67.61	24.47	(7826) ^a
Temperature	°C	40.00	25.85	22.80	29.99	25.00	25.00	25.00	29.99	25.00	25.00	25.00	29.99	25.00	25.00	25.00	29.99
Solid cont.	%	7.55	0.0067	0.2787	93.69	7.55	0.0049	0.3033	93.75	7.55	0.0022	0.2839	94.68	7.55	0.0004	0.3072	95.05
Moisture	%	–	–	–	4.17	–	–	–	3.14	–	–	–	3.39	–	–	–	3.15
NaCl	mg/L (%) ^b	1250	7.38	236	(1.94) ^b	1355	11.77	0	(2.90) ^b	1355	0.24	0	(1.72) ^b	1250	0	0	(1.60) ^b
COD	mg/L	63,034	66.13	2768	–	59,280	42.96	2654	–	59,280	17.20	2227	–	63,034	3.36	2315	–
TKN	mg/L	1325	0.92	38.57	–	1159	0.84	51.90	–	1159	0.34	43.48	–	1325	0.07	48.58	–
TN	mg/L	1445	1.15	48.01	–	1308	1.01	62.16	–	1308	0.38	49.13	–	1445	0.09	58.97	–
TP	mg/L	483	0.42	17.76	–	455	0.33	20.52	–	455	0.13	17.22	–	483	0.03	17.72	–

^a Those in parenthesis are the mass flow rates for whey powder while others in the row are the volumetric flow rates in the lines.

^b Those in parenthesis are the percent amounts (w/w) in whey powder while mg/L gives the concentration values in the lines.

Table 7
Overall economic results of whey treatment systems investigated.

Cost indicator	Unit	UF/RO	FO/MD		MD/RO			
			I ^a	II ^a	MD-PVDF		MD-PP	
					I ^a	II ^a	I ^a	II ^a
Whey powder production rate	Entity/year	103,225	104,493	104,493	103,494	103,494	103,099	103,099
Whey powder production cost	\$/entity	4.23	4.69	5.98	3.64	4.88	3.74	4.93
Operating cost	\$/year	437,000	490,000	625,000	377,000	506,000	386,000	508,000
Capital cost	\$	1,565,000	1,303,000	1,303,000	1,142,000	1,142,000	1,192,000	1,154,000
Revenue	\$/year	3,399,000	3,396,000	3,396,000	3,364,000	3,364,000	3,351,000	3,351,000
Return on investment	%	119.72	139.93	133.72	162.94	156.2	172.44	153.88
Pay-back time	years	0.84	0.71	0.75	0.61	0.64	0.58	0.65
NPV (at 7.0% interest)	\$	12,810,000	12,609,000	12,012,000	12,993,000	12,426,000	14,388,000	12,355,000
Cost of water produced	\$/m ³	14.23	11.25	17.33	6.70	12.61	6.59	12.33

^a I and II are the systems with and without heat recovery at MD process which means that energy sources were waste heat and electricity, respectively. Electrical energy consumptions in MD process with and without heat recovery were assumed as 0.52 and 23.20 kWh/m³, respectively [26].

decreased to 0.75/0.71, 0.64/0.61 and 0.65/0.58 year for FO/MD, MD (PVDF)/RO and MD (PP)/RO with/without heat recovery, respectively. Net present values were determined as 12,012,000/12,609,000, 12,426,000/12,993,000, and 12,355,000/14,388,000 \$ in the mentioned order. It is seen that somewhat high annual operating costs in innovative systems without heat recovery yielded partially lower NPVs between 384,000–798,000 \$, despite their lower capital costs compared to UF/RO. Although the use of waste heat in MD-included systems did not make NPV of FO/MD increase over that of UF/RO, it gives an excess of 183,000 and 1,578,000 \$ in NPVs of MD (PVDF)/RO and MD (PP)/RO, respectively. Unit costs of water produced at relative high values of 6.6–17.3 \$/m³ meant that water recovery from raw whey is to be economically viable with simultaneous mass recovery.

The economic performances of both FO/MD and MD/RO were determined as good as UF/RO, which is widely used in practice compared to others. It can be therefore declared that both innovative systems are economic alternatives to UF/RO. Although innovative scenarios were established to be as useful as the beneficial and profitable traditional practices, deficiency of knowledge and know-how at real case on design, operation and maintenance of innovative FO and MD processes have to be sensitively taken into consideration in order to make an apparent success in field application. More importantly, other important factors such as environment and society as well as technology and economics need to be considered carefully in evaluating industrial favourability of these systems. In that sense, future work will include a comparative technology assessment based on technical, economic

and environmental aspects. So, the most appropriate one(s) from five whey treatment scenarios including traditional UF/RO and innovative FO/RO (with NaCl draw solution), FO/T/RO (with NH₄HCO₃ draw solution including thermolysis (T) process), FO/MD and MD/RO membrane systems will be exposed in terms of their practical preferability.

4. Conclusions

As a sustainable alternative for effective pollution prevention in dairy industry, real-scale viability of two innovative whey treatment systems was techno-economically analyzed. Main findings of the research are (i) an increase in water recovery from 30% to 68% as well as the production of whey powder in commercial quality by using innovative systems, (ii) fast returns on investments with pay-back times as low as 0.84, 0.75 and 0.65 year for UF/RO, FO/MD and MD/RO, respectively, (iii) a return of 12–13 million \$ with an annual net profit of about 800,000 \$ for 15-year at 7% interest rate, and (iv) successful applicability of membrane technology for more sustainable dairy waste management.

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Nomenclature

COD	chemical oxygen demand (mg/L)
CTA	cellulose tri-acetate
CWTL	centralized wastewater treatment line
d	solution density (kg/L)
$\Delta\pi_{\text{net}}$	net osmotic pressure difference between draw and feed solutions in FO process (bar)
DS	draw solution in FO process
E_c	electrical conductivity (μ or mS/cm)
π_d	osmotic pressure of draw solution in FO process (bar)
π_f	osmotic pressure of feed solution in FO process (bar)
π_i	osmotic pressure of solution i in FO process (bar)
FO	forward osmosis
FS	feed solution in FO process
J	permeate flux in MD and RO processes ($\text{L}/\text{m}^2 \text{h}$)
J_w	water flux in FO process ($\text{L}/\text{m}^2 \text{h}$)
J_s	salt flux in FO process ($\text{g}/\text{m}^2 \text{h}$)
LFC	low fouling composite
m	osmolality (mosm/kg)
MD	membrane distillation
NF	nanofiltration
$\text{NH}_4\text{-N}$	ammonia-nitrogen (mg/L)
$\text{NO}_2\text{-N}$	nitrite-nitrogen (mg/L)
$\text{NO}_3\text{-N}$	nitrate-nitrogen (mg/L)
NPV	net present value
$\text{PO}_4\text{-P}$	orthophosphate-phosphorus (mg/L)
PP	polypropylene
PVDF	polyvinylidene fluoride
R	ideal gas constant ($8.314 \text{ J}/\text{K mol}$)
R	rejection ratio (%)
RO	reverse osmosis
RWL	raw whey line
SCOD	soluble chemical oxygen demand (mg/L)
SNF	fat-free dry matter (%)
t	time (h)
T	absolute temperature (K)
TKN	total Kjeldahl nitrogen (mg/L)
TN	total nitrogen (mg/L)
TP	total phosphorus (mg/L)
UF	ultrafiltration
V	permeated water volume (mL)
w	weight
W_{SC}	whey solid content (%)
WP	whey powder
WRL	water recovery line

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